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Radioactivity in Spanish spas

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Abstract

There are large number of spas in Spain and there is a lack of data concerning their radioactivity. The levels of radioactivity in a wide sample of Spanish spas were measured with special attention being paid to the radon and radium concentrations in the water, and to radon concentration in the indoor air of the spas. This study is primarily concerned with the radioactivity of the spas of one region in Spain — Cantabria — and particularly one spa where we measured radon levels in water as high as 824 Bq/l and over 5000 Bq/m^3 in the air of the rooms. We then considered a wider sample including virtually all of the radioactive spas in the country. The results indicate that a fairly large number of spas have radon levels in water that are moderately high and they are used for the treatment of diseases without radiation protection measures for patients or staff at the spas.

Keywords: Radon; Radium; Water; Air; Cantabria

1. Introduction

At present there are almost 100 places in Spain used as health spas where a large and growing number of patients are treated. Some of these spas have a long history going back to their use as baths in the times of the Romans or occupation under the Moors, but most of them were built towards the end of the nineteenth century. While spas are widely spread throughout Spain, they tend to occur, as do the springs on which they depend, in mountainous areas.

Most of the spa waters that exist in Spain have been analysed in detail and their chemical properties and physical properties such as the temperature of the emergent water have been published [1]. However, other properties such as the concentration of radioactive elements in the water are not known except for measurements made some years ago with unreliable portable equipment [2]. The aim of this research is to attempt to characterize the level of radioactivity present in Spanish spas. Measurements were made of the concentration of ^{222}Rn dissolved in the water of a wide range of spas selected from the most radioactive according to a previous study, and the concentration of ^{226}Ra and the radon

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concentration in the air used for inhalation treatment. In some spas inhalation facilities have been installed and aerosols can be generated by dispersion of mineral water with varying size distributions. The air inhaled by the patients can contain radon and its decay products with an equilibrium factor in the range 0.15–0.30.

2. Methods

Measurement of the radioactive elements dissolved in the water was made from samples taken from 54 spas throughout the whole of Spain from 1989 to 1991. The water samples were taken at the source of the spring water, or as close as possible to this point, using 0.8-l hermetically sealing lucite samplers with Marinelli geometry adapted to the dimensions of the detector.

The concentration of the radon dissolved in water was measured with a 76 mm × 76 mm NaI(Tl) scintillation detector protected from external radiation by a lead shield and connected to a multichannel analyser. Each sample was measured for 20 000 s at least 3 h after collection. The system was calibrated with ^{226}Ra solutions of known activity in equilibrium with its decay and prepared with the same geometry as the samples to be measured. Under the experimental conditions used for counting time, sample geometry, background noise and correction for radioactive decay, the detection limit of the method is 4 Bq/l for radon and the measurement error does not exceed 8% of the measured value [3].

Measurement of the concentration of ^{226}Ra was performed later using the same samples. The method followed in this case involved radiochemical separation of the radium with inactive barium as carrier until a solid sample was deposited onto a stainless steel planchet [4]. Three days after preparation the sample thus obtained was counted for 60 000 s with an ZnS α-radiation detector coupled to a photomultiplier linked to a scaler. In the experimental conditions used, the detection limit of the method is 4 mBq/l for the ^{226}Ra concentration and the error does not exceed 10% of the value found.

The concentration of radon in the air of spas both in inhalation systems and in the atmosphere was measured with 1-l lucite cells in which a vacuum had been created. The cells were lined with a sheet of ZnS and were counted with a system composed of a photomultiplier, amplifier and counter for 20 min 3 h after collection. The detection limit in these conditions is 10 Bq/m^3 and the estimated error 10% of the measured concentration [5].

3. Results

Cantabria, a region situated in the north of Spain, has a long tradition in spas and possesses a large number of springs. In this region we made a series of measurements of the water of ten spas in order to determine the level of dissolved radon and the possible seasonal variation in concentration levels. Samples of the spa water were, therefore, taken every 2 months during a year and measurement was then made of the radon and radium concentrations in the samples. Deviations of the concentrations measured for each individual spa compared with the annual mean for each spa are below 10% in all cases and, therefore, of the same order of magnitude of the experimental errors associated to the measurement method.

The radon concentrations found vary considerably from spa to spa and range from < 4 Bq/l to 840 Bq/l. No definite relationship was observed between radon concentration and such other characteristics of the water as chemical composition or temperature of the emergent water.

The concentrations of radium dissolved in the water range from < 4 mBq/l to 860 mBq/l. As with most deep waters there is a great difference between the radium and radon concentrations in the water. The concentrations of radium found, as in the case of radon, do not readily correlate with other chemical characteristics of the water or with the spring water temperature. In contrast, there is a degree of correlation between radon concentration and radium concentration. Although the relationship between the two is not directly proportional, high radon values correspond to relatively high radium values.

In our study, special interest was shown in the spa of 'Las Caldas de Besaya' situated in the region of Cantabria. It has seven springs although

only the one with the largest volume has important radon concentrations in the waters. The concentrations were measured over a whole year and a constant value of 824 ± 51 Bq/l was found. The water from this spring flows through pipes to the baths located at the point of emergence, where the radon concentration measured was 560 ± 80 Bq/l.

The radon concentration in the air inside the spa was measured at different points including points close to the spring, and inside the bath and shower rooms and the inhalation room. Measurements were also taken in the entrance hall situated on the upper floor. The values found ranged from 3560 and 6650 Bq/m^3 for the various points in the baths. In the entrance hall the values were lower with an average of 580 Bq/m^3. In all cases the concentration measured is far higher than the average value found inside Spanish buildings [6], which is ~ 40 Bq/m^3.

The above data allow us to make an estimation of the relationship between the radon concentrations in water and those in air inside the spa. Thus, given that the flow is 300 l/min and the concentration in the water is 560 Bq/l, radon generation inside the spa will be 4.68×10^6 Bq/h. If we assume a ventilation rate of 1 h^{-1}, given the volume 1000 m^3 of the room, this rate of radon generation will result in a radon concentration in the air of 4680 Bq/m^3, which agrees very well with the measured values.

Further measurements were made of the radon concentration in the air used in inhalation treatment. This air comes directly from the source of the spring and is breathed in by patients through inhalation tubes in order to take advantage of the gases dissolved in the water. The radon concentration measured from air samples taken directly from the inhalers is between 99400 and 102700 Bq/m^3. With this radon concentration, in a treatment of 10 h over a period of 20 days per year, the patients are exposed to dose equivalent to bronchial epithelium of 33 mSv/year and an effective dose equivalent to total body for inhalation of 4 mSv/year, double the effective dose equivalent due to exposure to natural sources of radiation, established to be 2 mSv/year.

Both the radon concentrations in the water and those in the inhalation air, but especially the latter, show values high enough for the spa particularly favourable for use in the development of research studies concerning the health effects and the risk/benefit ratio of spa water treatment with high ^{222}Rn concentrations. However, it should be noted that the high values in the ambient air represent a risk to the staff at the spa [7,8]. For the evaluation of the annual effective dose equivalent to the staff, we have considered a working period of 10 weeks per year (because the spa only opens for the summer months), a mean radon concentration in air of 5000 Bq/m^3, and an environmental-conversion factor of 0.007 Gy/WLM for bronchial epithelium. The estimated doses, 200 mSv/year to the bronchial epithelium and 24 mSv/year to the total body, are similar to the maximum allowable effective dose equivalent proposed by the ICRP N60 (1991) — 50 mSv in any single year or 100 mSv in any 5-year period — for occupational exposure limits.

Parallel to the above measurements, we carried out a survey of the radioactivity in the water at a set of another 44 spas throughout Spain. The approximate location of the spas included in the survey is shown in Fig. 1. At least two water samples were taken at each site and both radon and radium concentrations were measured.

Table 1 shows the results obtained for the different spas studied, classifying them in five groups by their geographical location showing the ranges for ^{222}Rn and ^{226}Ra concentrations in water as well as ^{222}Rn in air.

In order to describe the concentrations of radon dissolved in water we divided the measured values by intervals as shown in Fig. 2, where the values for Cantabria are also included. It can be seen that there are 25 spas with radon concentrations < 20 Bq/l; 15 with concentrations between 20 and 150 Bq/l; seven with concentrations between 150 and 300 Bq/l; and seven with higher concentrations. Only the latter can strictly speaking be considered radioactive, for therapeutical purposes, according to the legislation applied in other countries [2,8]. For this last group, the radon concentration in water is similar to that existing

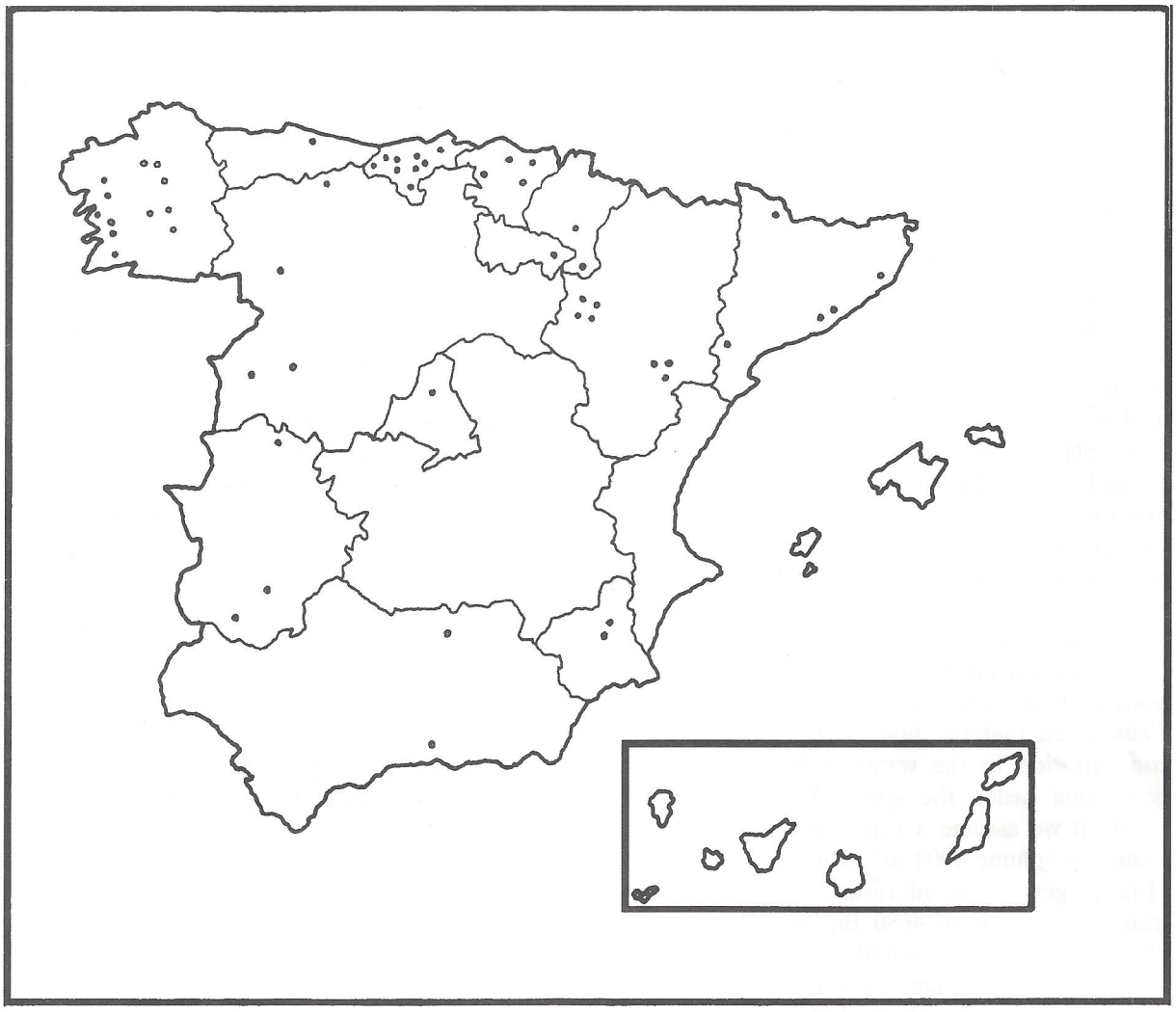

Fig. 1. Location of the spas studied in the country.

Table 1
Concentrations of the ^{222}Rn and ^{226}Ra in water and ^{222}Rn in the air for the samples collected from the spas

Area	No of spas	No of samples	Range of ^{222}Rn in water (Bq/l)	Range of ^{222}Rn in air (Bq/m^3)	Range of ^{226}Ra in water (mBq/l)
Northwest	12	32	[a]L.D.–402	L.D.–1260	L.D.–3660
Cantabria	10	56	L.D.–840	L.D.–5000	L.D.–860
North (other)	10	25	L.D.–595	L.D.–940	L.D.–120
Northeast	12	27	L.D.–614	L.D.–240	L.D.–660
Spain (other)	10	24	L.D.–342	L.D.–310	L.D.–270

[a]L.D., detection limit.

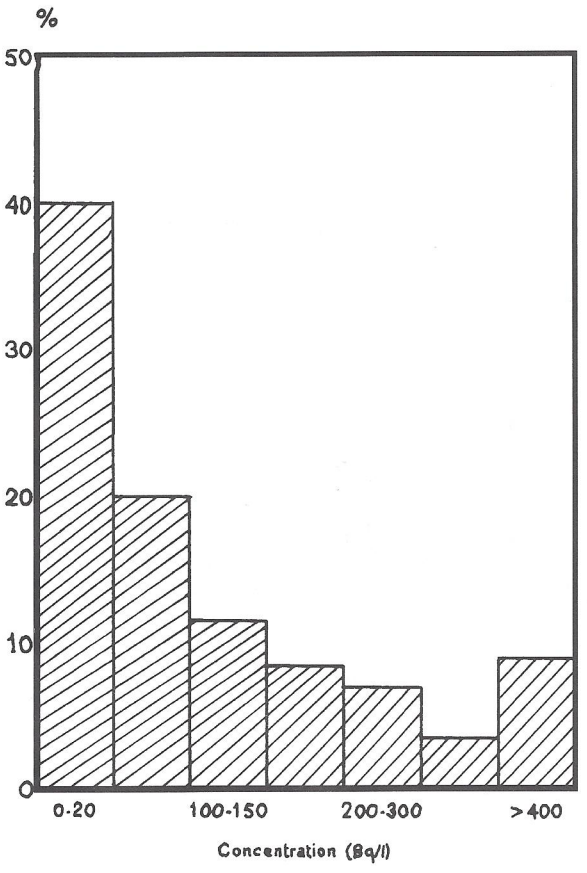

Fig. 2. Percentages of spas versus the radon in water measured.

in other locations where radon treatment is performed [9], and higher than others considered radioactive [10,11].

The seven most radioactive spas (over 300 Bq/l) are geographically situated in such a way that the correlation with the radioactive characteristics of the local geology is far from simple; they are not located in those areas of the country where granitic and shales formations are present and where radium concentrations in soils and rocks are highest. Of the seven spas referred to above, five are situated in the north of the Iberian Peninsula along the chain of the Cantabrian Mountains and the Pyrenees, where very diverse geological formations occur with rocks possessing a wide range of values for the radium concentration.

In the Cantabrian spas the radon concentration in spa water does not correlate with spring water temperature. In the very radioactive group there are springs with a temperature range from 28°C to 61°C, which is similar to that of springs showing lower levels of radon in water.

The concentration of radium in the waters of the spas studied varies from values below our lower detection limit of 4 mBq/l to values as high as 3660 mBq/l. The results are approximately lognormally distributed, and the calculated geometric mean is 26 mBq/l.

A considerable number of spas show ^{226}Ra water concentrations < 20 mBq/l, and there is another group of 11 spas which concentrations exceeded the currently accepted maximum contaminant level (MCL) of 185 mBq/l.

At 12 of the spas we determined the value of the radon concentration in the air both for ambient air and that in the inhalers or inhalation rooms. Of the 12 spas where radon concentration in air was measured, three had low radon concentration in water. The air values for these were very low, similar to those found in the houses in the surrounding areas. The other nine had high or very high concentrations in water. The radon concentration in the air of these spas was always higher than that of the houses of the area but without reaching very high values, below 450 Bq/m^3, except in one case.

By contrast, the air used for inhalation treatment showed higher concentrations ranging from 4200 to 102 000 Bq/m^3. These high values, which depend on the concentration in water, flow and even the type of installation, correlate only weakly with the radon concentration in water.

4. Conclusions

A broad sample of values has been achieved for the radioactive content in water and air at spas throughout Spain. Special care was taken to include in this sample the spas where radioactivity was supposed to be high.

Of the spas measured in the survey seven had

high values for radon in water and another seven had moderately high values. The radon concentration in the water in spas does not show a simple correlation with local geology, with the ^{226}Ra concentration, or with water temperature.

The radon concentration in ambient air of the radioactive spas is moderately high, except in one case. The radon concentration in the air of inhalers and inhalation rooms is, however, much higher.

Finally, we consider that the spa at 'Las Caldas de Besaya' (Cantabria) possesses characteristics for radon concentration in water and in inhalation air that make it particularly suited for the development in a close future of more specific studies concerning health effects of spa water treatment with high ^{222}Rn radon concentrations.

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